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Escape-rate formalism, decay to steady states, and divergences in the entropy-production rate

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In summer 1997 we were sitting with Bob Dorfman and a few other friends interested in chaotic systems and transport theory on a terrace close to Oktogon in Budapest. While taking our (decaf) coffee after a very nice Italian meal, we discussed about logarithmic divergences in the entropy production of systems with absorbing boundary conditions and their consequences for the escape-rate formalism. It was guessed at that time that the problem could be resolved by a careful discussion of the *physical* content of the absorbing boundary conditions. To our knowledge a thorough analysis of this long-standing question is still missing. We dedicate it hereby to Bob on occasion of his 65th birthday.

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I. INTRODUCTION

The *escape-rate formalism* [1, 2, 3, 4] aims at identifying transport coefficients based on the asymptotic decay rate of an initial non-stationary density profile towards an empty steady state selected by absorbing boundary conditions where all particles disappear from the interior of the system. In this formulation the relaxation problem has widely been studied in the context of Markov chains (*cf.* the sections on *survival probabilities* in [5], and on *absorbing states* in [6]), as well as for deterministic chaotic systems [7, 8, 9]. However, the choice of an empty asymptotic state places severe constraints on the formalism. In particular, the thermodynamic entropy

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production picks up logarithmically diverging contributions at the boundaries. After all, a vanishing density is physically unrealistic. In particular, it leads to the breakdown of the concept of an entropy-production density even in the framework of classical irreversible thermodynamics. Close to a boundary at $x = 0$ the absorbing boundary condition requires a density profile of the form $\rho(x) = \alpha x$. For purely diffusive particle transport, a neighborhood of size Δ gives then rise to the entropy production¹

$$\Sigma^{(\text{irr})}(\Delta) \equiv \int_0^{0+\Delta} dx \, D \left(\frac{\partial_x \rho}{\rho} \right)^2 \rho = D\alpha \lim_{\delta \rightarrow 0} \ln \frac{\Delta}{\delta}. \quad (1)$$

In the present paper this logarithmic divergence will be discussed from the point of view of spatially extended chaotic systems whose transport properties fully agree with the predictions of irreversible thermodynamics. To keep the calculations as transparent as possible, the discussion is given for isothermal multibaker maps. However, the described picture should apply in general.

In Sect. II Eq. (1) is contrasted with the prediction of the irreversible entropy changes in an entropy based on the conditional density characterizing the chaotic saddle forming the backbone of transport in the system. This prediction always yields finite values. The isothermal multibaker map is introduced in Sect. III, where also its entropy balance is worked out. Sect. IV deals with the normal modes of the coarse-grained time evolution. This allows us to address in Sect. V the origin of the logarithmic divergences in the entropy production from the point of view of a microscopic reversible dynamics. The analysis makes use of the eigenvalues of the time-evolution operator [4, 10], which does not depend on the nature of the asymptotic state. To underline this observation, we also discuss asymptotic states with uniform nonzero densities, and point out how the divergences are lifted by an arbitrarily small background density. The discussion also shows why the changes of the irreversible entropy based on the conditional density differ from the thermodynamic expectation. The presence of an arbitrarily small background density in the asymptotic case turns the entropy production to be finite, but different from the prediction based on the conditional density. In the concluding Sect. VI these findings are complemented by a discussion of the behavior of the entropy production in systems relaxing towards a typical nonempty steady state of finite density as compared to the case of small (or even vanishing) background densities addressed in the main part.

¹ The Boltzmann constant is taken to be unity throughout this paper.

II. ENTROPY PRODUCTION BASED ON CONDITIONAL INVARIANT MEASURES REVISITED

One of the early studies of thermodynamic entropy production in deterministic dynamical systems was based on the escape-rate formalism introduced by Gaspard and Nicolis [1]. As a generalization of it, Breymann, Tél and Vollmer [3] considered open dissipative dynamical systems in continuous time. To characterize their irreversible features, they suggested to use the entropy

$$s(t) = - \int dx \psi^{(t)}(x) \ln \psi^{(t)}(x) \quad (2)$$

based on the *normalized conditional phase-space density* $\psi^{(t)}(x)$, describing the probability to find a point which has not yet escaped the system by time t at phase-space coordinate x . Because $\psi^{(t)}(x)$ is a single-particle property, $s(t)$ can be considered as a specific entropy (total entropy per number of particles). The initial condition is selected from an arbitrary *smooth* distribution $\psi^{(0)}(x)$. As time goes on, the phase-space volume of $\psi^{(t)}$ is exponentially shrinking as $\exp[-\zeta(x)t]$, where the local phase-space contraction rate $\zeta(x) = \sum \lambda_i(x) - \kappa$ is a smooth function of the coordinates [7]. Here, κ is the escape rate from the system, and $\lambda_i(x)$ are the local Lyapunov exponents that describe the phase-space contraction in the independent directions i in phase space (*cf.* [7, 8] for detailed discussions of conditional invariant densities and Lyapunov exponents). On its support the value of the conditional density $\psi^{(t)}(x)$ is exponentially increasing due to its normalization. More precisely, it increases like $\psi^{(t+dt)}(x) = \exp[\zeta(x)dt] \psi^{(t)}(x) \chi^{(t+dt)}(x)$, where $\chi^{(t+dt)}$ is the characteristic function of the support at time $t+dt$. The entropy $s(t+dt)$ at time $t+dt$ can be determined by inserting this relation into Eq. (2):

$$\begin{aligned} s(t+dt) &= -dt \int dx \zeta(x) \psi^{(t)}(x) e^{\zeta(x)dt} \chi^{(t+dt)}(x) \\ &\quad - \int dx \psi^{(t)}(x) \ln \left[\psi^{(t)}(x) \chi^{(t+dt)}(x) \right] e^{\zeta(x)dt} \chi^{(t+dt)}(x). \end{aligned} \quad (3)$$

In both integrals the decrease of the support of ψ is counterbalanced by the factor $\exp[\zeta(x)dt]$. The first integral is the phase-space average $\bar{\zeta}$ of $\zeta(x)$. For $dt \rightarrow 0$ the second one tends to the specific entropy $s(t)$ at time t . Hence, in the long-time limit the time derivative of the entropy

$$\frac{ds}{dt} = -\bar{\zeta} = \sum_i \bar{\lambda}_i - \kappa, \quad (4)$$

is the difference of the sum of the average Lyapunov exponents $\bar{\lambda}_i$ on the saddle, and the escape rate κ from the saddle. The average is taken with

respect to the density $\psi(x)$ of the conditionally-invariant measure. This measure is time independent. Its support is the unstable manifold of the chaotic saddle, *i.e.*, the union of the never escaping orbits in the system. The fact that the time derivative approaches a constant reflects the ever refining fractal structures in the density due to the chaoticity of the dynamics.

We now compare $s(t)$ with a coarse-grained entropy $s^{(\text{cg})}(t)$ computed in an analogous way from a coarse-grained conditional density $\psi^{(\text{cg})}(t)$, which — in contrast to ψ — does converge towards a stationary distribution. The irreversible entropy production is then obtained as (*cf.* [3])

$$P^{(\text{irr})} \equiv \frac{d}{dt} \left[s^{(\text{cg})}(t) - s(t) \right] \xrightarrow{\text{long times}} \kappa - \sum_i \bar{\lambda}_i. \quad (5)$$

It measures the lack of information on the microscopic state due to the finite resolution of the coarse-grained description. Similarly to s , $P^{(\text{irr})}$ is a specific quantity.

In systems with a reversible dynamics the phase-space contraction is proportional to the displacement in the direction of an applied field [11, 12, 13, 14, 15, 16]. Consequently, in an open system with reversible dynamics the sum of the average Lyapunov exponents on the chaotic saddle is zero since the average number of steps in the direction of positive phase-space contraction is the same as in the opposite. Therefore, its specific irreversible entropy production amounts to the escape rate,

$$P^{(\text{irr})} = \kappa. \quad (6)$$

In the following we revisit this argument in the light of recent developments [13, 17] dealing with steady states instead of empty asymptotic states. We work out the irreversible entropy production for an isothermal multibaker map with reversible microscopic dynamics subjected to absorbing boundary conditions.

III. THE ISOTHERMAL MULTIBAKER MAP

Multibaker maps model particle transport in spatially extended systems by a chain of mutually interrelated baker maps [13, 14, 17, 18, 19, 20, 21, 22, 23, 24]. They consist of N identical cells of width a and height 1 (the phase-space) in the (x, p) plane. The cells are labeled by the index m (Fig. 1a). After each time unit τ , every cell is divided into three columns (Fig. 1b). Here we consider the case when the right (left) column of width ar (al) is mapped onto a strip of width a and of height l (r) in the right (left) neighboring cell. The middle one, which is of width as , preserves its area, such that its image attains a height s , and $r + l + s = 1$. There are

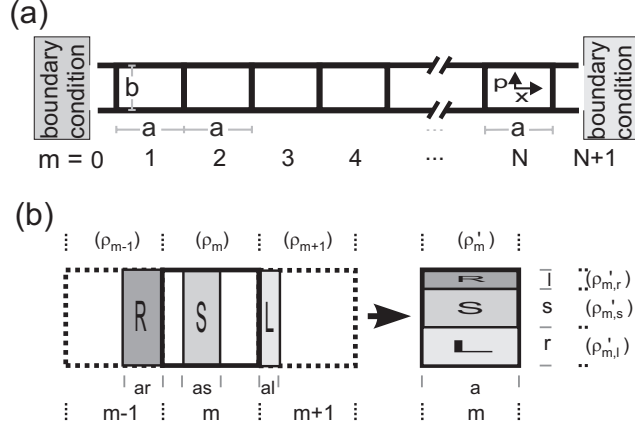


FIG. 1: Graphical illustration of the action of the multibaker map of length $L = aN$ on the phase space (x, p) over a time unit τ . (a) The mapping is defined on a domain of N identical rectangular cells of size $a \times 1$, with boundary condition imposed in two additional cells 0 and $N + 1$. (b) The action of the map in any of the cells over time unit τ is illustrated by the deformation of the labels R , S and L in the three branches of the map ending up in cell m . The average value of the density on the cells (strips) [*cf.* Eq. (7)] is given on the margins.

more general parameter settings conceivable, but earlier work [13, 16, 17] showed that the associated macroscopic behavior is then not compatible with irreversible thermodynamics.

The dynamics of the multibaker map models a microscopic dynamics described in the single-particle phase space. It is deterministic, invertible, chaotic, and mixing [15, 25]. To describe irreversible processes one follows the coarse-grained densities ρ_m obtained by averaging over the cells [13, 14, 17]. To emphasize the particular choice of coarse-graining over the cells, the coarse-grained densities are also called the *cell densities*. The dynamics of the multibaker map is the same for all cells. There might be inhomogeneities in the densities, but the evolution equations are translation invariant.

A. Evolution of the cell density

In order to find results consistent with non-equilibrium thermodynamics we always consider initial conditions with a uniform density in every cell m . This is convenient from a technical point of view, and does not lead to a principal restriction of the domain of validity of the model as discussed

in [13, 16, 26]. Under such conditions the parameters r and l can be considered as transition probabilities from a cell to its right and left neighbor, respectively. After one step of iteration the densities $\rho'_{m,i}$ on the strips $i = R, S, L$ of cell m are (*cf.* Fig. 1b)

$$\rho'_{m,r} = \frac{r}{l} \rho_{m-1}, \quad \rho'_{m,s} = \rho_m, \quad \rho'_{m,l} = \frac{l}{r} \rho_{m+1}. \quad (7)$$

The factors r/l and l/r give rise to local contraction or expansion of the phase-space volume. One of the factors is larger than unity, and characterizes a local contraction, while the other gives rise to an expansion.

Since in every cell the density remains uniform in the horizontal direction, this update holds at all times such that the coarse-grained density distributions ρ_m and $\rho'_m = r\rho_{m-1} + s\rho_m + l\rho_{m+1}$ at the respective times n and $n+1$ are related by the master equation

$$\rho'_m = (1 - r - l)\rho_m + r\rho_{m-1} + l\rho_{m+1}. \quad (8)$$

Multiplying the equation by τ^{-1} and introducing the current

$$j_m = \frac{a}{\tau}(r\rho_m - l\rho_{m+1}) \quad (9)$$

through the right boundary of cell m , Eq. (8) appears in the form of the continuity equation

$$\frac{\rho'_m - \rho_m}{\tau} = -\frac{j_m - j_{m-1}}{a}. \quad (10)$$

The current through the left boundary of cell m is the same as the current flowing through the right boundary of cell $m-1$.

B. Diffusion, drift, and the macroscopic limit

The transition probabilities r and l govern the evolution of the coarse-grained density ρ_m . In view of the master equation (8), the cell-to-cell dynamics of the model is equivalent to the dynamics of an ensemble of random walkers with fixed step length a and local transition probabilities r and l over time unit τ . In terms of the local drift v and diffusion coefficient D [27] the transition probabilities r and l can be expressed as

$$r = \frac{\tau D}{a^2} \left(1 + \frac{av}{2D}\right), \quad l = \frac{\tau D}{a^2} \left(1 - \frac{av}{2D}\right), \quad (11)$$

such that the current appears in a form very close to its thermodynamic counterpart, viz.

$$j_m = \frac{v}{2}(\rho_m + \rho_{m+1}) - D \frac{\rho_{m+1} - \rho_m}{a}. \quad (12)$$

The *macroscopic limit* expresses a separation of scales where density gradients inside cells may be neglected, while density differences between the cells and the temporal evolution of the cell densities are only taken into account in leading order. In other words, in this limit $a \ll L = aN$, $\tau \ll L^2/D$, L/v , and upon introducing the quasi-continuous spatial and temporal variables $x \equiv am$ and $t \equiv \tau n$, the current j_m [Eq. (12)] takes the macroscopic limit $j(x; t) = v \rho(x; t) - D \partial_x \rho(x, t)$, while Eq. (10) becomes $\partial_t \rho(x; t) = -\partial_x j(x; t) = -v \partial_x \rho(x; t) + D \partial_x^2 \rho(x; t)$. The macroscopic density evolves according to the advection-diffusion equation.

C. Local entropy balance

The coarse-grained entropy of cell m is defined as

$$S_m = -a \rho_m \ln \frac{\rho_m}{\rho^*}. \quad (13)$$

It fulfills a local entropy balance in direct analogy to the one of irreversible thermodynamics [13, 14]. In this equation ρ^* is a constant reference density that is introduced for dimensional reasons. In classical physics it expresses the free choice of the origin of the entropy scale.

To derive the balance equation for (13) one identifies at any given time the difference $S_m - S_m^{(G)}$ of the coarse-grained and the Gibbs entropy $S_m^{(G)}$ as the information on the microscopic state of the system which cannot be resolved in the coarse-grained description. The Gibbs entropy is an analogous expression to (13) given in terms of the non-coarse-grained phase-space density ρ , as $-\rho \ln \rho / \rho^*$ integrated over the cell. For a coarse-grained initial distributions the entropies coincide initially (*i.e.*, $S_m = S_m^{(G)}$). After one time step the entropies become (*cf.* Fig. 1b)

$$S_m^{(G)'} = -a \left[s \rho_m \ln \frac{\rho_m}{\rho^*} + r \rho_{m-1} \ln \left(\frac{r}{l} \frac{\rho_{m-1}}{\rho^*} \right) + l \rho_{m+1} \ln \left(\frac{l}{r} \frac{\rho_{m+1}}{\rho^*} \right) \right] \quad (14)$$

and

$$S'_m = -a \rho'_m \ln \frac{\rho'_m}{\rho^*}. \quad (15)$$

The temporal change of the lack of information is identified with the irreversible entropy production $\Delta_i S_m$, and the change $(S_m^{(G)'} - S_m^{(G)})$ of the Gibbs entropy with the entropy flux $\Delta_e S_m$. Thus, one obtains the discrete entropy balance in any cell

$$\frac{S'_m - S_m}{\tau} = \frac{\Delta_e S_m}{\tau} + \frac{\Delta_i S_m}{\tau}. \quad (16)$$

The form of the entropy production is [cf. (14) and (15)]

$$\begin{aligned} \frac{\Delta_i S_m}{\tau} &= \frac{[S'_m - S_m^{(G)'}] - [S_m - S_m^{(G)}]}{\tau} \\ &= \frac{a}{\tau} \left[-\rho'_m \ln \frac{\rho'_m}{\rho_m} + r \rho_{m-1} \ln \left(\frac{r}{l} \frac{\rho_{m-1}}{\rho_m} \right) + l \rho_{m+1} \ln \left(\frac{l}{r} \frac{\rho_{m+1}}{\rho_m} \right) \right], \end{aligned} \quad (17)$$

and the entropy flux becomes

$$\begin{aligned} \frac{\Delta_e S_m}{\tau} &= \frac{S_m^{(G)'} - S_m^{(G)}}{\tau} \\ &= -\frac{a}{\tau} \left[(\rho'_m - \rho_m) \ln \frac{\rho_m}{\rho^*} + r \rho_{m-1} \ln \left(\frac{r}{l} \frac{\rho_{m-1}}{\rho_m} \right) + l \rho_{m+1} \ln \left(\frac{l}{r} \frac{\rho_{m+1}}{\rho_m} \right) \right] \end{aligned} \quad (18)$$

In the macroscopic limit all expressions reduce to the respective predictions of non-equilibrium thermodynamics [13, 14, 16]. Note that the entropy production does not depend on the choice of the reference density ρ^* . The density of irreversible entropy production is then $\Delta_i S_m / (a\tau)$.

D. Global entropy production

The escape-rate formalism addresses the balance of the global entropy of the chain. The global coarse-grained entropy is

$$S_{\text{tot}} = \sum_{m=1}^N S_m = -a \sum_{m=1}^N \rho_m \ln \frac{\rho_m}{\rho^*}. \quad (19)$$

The associated global entropy production rate along the chain is $\Sigma^{(\text{irr})} = \sum_{m=1}^N \Delta_i S_m / \tau$, and the total specific irreversible entropy production is obtained as

$$P^{(\text{irr})} = \frac{\Sigma^{(\text{irr})}}{\mathcal{N}} = \frac{\sum_{m=1}^N \Delta_i S_m}{\tau \mathcal{N}} \quad (20)$$

where $\mathcal{N} = \sum_{m=1}^N a \rho_m$ is the number of particles in the chain at time $n\tau$. The total entropy production can be rearranged to take the form

$$\begin{aligned} \Sigma^{(\text{irr})} &= -\frac{a}{\tau} \sum_{m=1}^N \rho'_m \ln \frac{\rho'_m}{\rho_m} + \frac{a}{\tau} \left(r \rho_0 \ln \frac{r \rho_0}{l \rho_1} - l \rho_{N+1} \ln \frac{r \rho_N}{l \rho_{N+1}} \right) \\ &\quad + \frac{a}{\tau} \sum_{m=1}^{N-1} (r \rho_m - l \rho_{m+1}) \ln \frac{r \rho_m}{l \rho_{m+1}}, \end{aligned} \quad (21)$$

where ρ_0 and ρ_{N+1} are the densities in the boundary cells. It can conveniently be split into four terms $\Sigma^{(\text{irr})} = \Sigma_t^{(\text{irr})} + \Sigma_b^{(\text{irr})} + \Sigma_d^{(\text{irr})} + \Sigma_{\text{mix}}^{(\text{irr})}$. The first one

$$\Sigma_t^{(\text{irr})} = -\frac{a}{\tau} \sum_{m=1}^N \rho'_m \ln \frac{\rho'_m}{\rho_m} \quad (22)$$

is the contribution from the *temporal* evolution of the density. The contribution proportional to $\ln(r/l)$ of the last term in (21) contains the irreversible entropy production

$$\Sigma_d^{(\text{irr})} = \mathcal{N} \frac{r-l}{\tau} \ln \frac{r}{l} \rightarrow \mathcal{N} \frac{v^2}{D} \quad (23)$$

due to the presence of the *drift* v . It does not depend on the particular density distribution so that we could immediately specify its macroscopic limit (indicated by \rightarrow). By means of (9) and (10) the remaining part can be written as a sum of two terms. One of them,

$$\Sigma_{\text{mix}}^{(\text{irr})} = -\frac{a}{\tau} \sum_{m=1}^N (\rho'_m - \rho_m) \ln \frac{\rho_m}{\rho^*} \quad (24)$$

characterizes the contribution of *mixing* of the neighboring densities. In order to arrive at this form the ratio ρ_m/ρ_{m+1} of the densities appearing at the right hand side of (21) was written as $[(\rho_m/\rho^*)/(\rho_{m+1}/\rho^*)]$. The rest

$$\Sigma_b^{(\text{irr})} = \frac{a}{\tau} \left[r \rho_0 \ln \frac{r \rho_0}{l \rho^*} + l \rho_{N+1} \ln \frac{l \rho_{N+1}}{r \rho^*} - l \rho_1 \ln \frac{l \rho_1}{r \rho^*} - r \rho_N \ln \frac{r \rho_N}{l \rho^*} \right] \quad (25)$$

yields the *boundary* contribution. We shall be interested in the difference between $\Sigma^{(\text{irr})}$ and $\Sigma_d^{(\text{irr})}$, called the irreversible entropy production $\Sigma_{\text{relax}}^{(\text{irr})}$ connected to the *relaxation* process,

$$\Sigma_{\text{relax}}^{(\text{irr})} \equiv \Sigma_t^{(\text{irr})} + \Sigma_{\text{mix}}^{(\text{irr})} + \Sigma_b^{(\text{irr})}. \quad (26)$$

IV. NORMAL MODES OF THE COARSE-GRAINED TIME EVOLUTION

A. Decaying modes and the steady state

We are interested in the evolution of the density distributions $\rho_m^{(n)}$ subjected to a fixed constant boundary condition

$$\rho_0^{(n)} = \rho_{N+1}^{(n)} = \rho_B \quad (27)$$

at any time step n . Asymptotically, $\rho_m^{(n)}$ always approaches the uniform density ρ_B . The time evolution of the density can be explored by expanding the deviation $\rho_m^{(0)} - \rho_B$ of the initial distribution from the asymptotic state in terms of normal modes $\delta_m^{[\nu](n)}$. They vanish at both boundaries, and only change in amplitude but not in their shape,

$$\frac{\delta_m^{[\nu](n+1)}}{\delta_m^{[\nu](n)}} = \exp(-\gamma_\nu \tau). \quad (28)$$

The integer ν labels different modes. There are as many independent modes as the number N of the cells $\nu = 1, \dots, N$. The normal modes take the respective forms

$$\delta_m^{[\nu](n)} \sim \exp(-\gamma_\nu n \tau) \left(\frac{r}{l}\right)^{m/2} \sin\left(\frac{\pi \nu}{N+1} m\right). \quad (29)$$

Substituting the ansatz into Eq. (8) and rearranging the trigonometric terms, one finds the decay rates

$$\gamma_\nu = -\frac{1}{\tau} \ln \left[1 - (r+l) + 2\sqrt{rl} \cos\left(\frac{\pi \nu}{N+1}\right) \right] \rightarrow \frac{\pi^2 D}{L^2} \nu^2 + \frac{v^2}{4D}. \quad (30)$$

For a general initial condition the *asymptotic* decay is governed by the *slowest* non-vanishing decay rate, γ_1 . It coincides with the *escape rate* κ of the transiently chaotic motion [8] inside the chain (i.e., $\kappa \equiv \gamma_1$) [1, 20, 28].

The macroscopic limit of the decay rates has a clear physical content. For vanishing v it states that relaxation is related to the typical diffusive decay rate D/L^2 of structures of size L . The factor π^2 characterizes the geometry of the considered region (a band of width L with straight, parallel walls in our example). More complicated geometries have been studied recently by Gaspard [18], and Kaufmann and collaborators [29, 30].

For a biased motion $v \neq 0$ the drift singles out one side of the system and sweeps out the particles in that direction. This mechanism dominates when the time L/v to cross the system by the biased motion becomes shorter than the typical time scale L^2/D of diffusion, *i.e.*, for

$$\text{Pe} \equiv \left| \frac{vL}{D} \right| \quad (31)$$

much larger than unity. In the context of hydrodynamics, Pe is called Péclet number [31]. It measures the importance of diffusion relative to advection. Strong diffusive effects are indicated by small Péclet numbers. For fixed finite v and D , the Péclet number is always large for a sufficiently large system size L .

B. Long-time relaxation and the slowest mode

For sufficiently long times $n \gg 1$, the coarse-grained density closely approaches the first normal mode. Therefore, the density can be expressed as

$$\rho_m^{(n)} \equiv \rho_B + \left(\mathcal{N}^{(n)} - \mathcal{N}^{(\infty)} \right) \psi_m, \quad (32)$$

where $\mathcal{N}^{(\infty)} = \rho_B L$ is the particle number in the background which is also the asymptotic particle number in the system, and

$$\psi_m = \frac{\mathcal{A}}{L} \left(\frac{r}{l} \right)^{m/2 - (N+1)/4} \sin \frac{m\pi}{N+1} \quad (33)$$

is the *coarse-grained* conditionally-invariant density. It is normalized to unity ($1 \equiv a \sum_{m=1}^N \psi_m$), by virtue of the normalization constant \mathcal{A} , which is invariant under the exchange of r and l . Carrying out the summation of the complex geometric series defined by (33) one finds

$$\begin{aligned} \mathcal{A} &= L \frac{1 - \exp(-\kappa\tau)}{a\sqrt{rl}} \frac{1}{\left(\frac{r}{l}\right)^{(N+1)/4} + \left(\frac{l}{r}\right)^{(N+1)/4}} \frac{1}{\sin \frac{\pi}{N+1}} \\ &\rightarrow \frac{\pi}{2} \frac{\left(\frac{\text{Pe}}{2\pi}\right)^2 + 1}{\cosh \frac{\text{Pe}}{4}} = \kappa \frac{L^2}{2\pi D \cosh \frac{\text{Pe}}{4}}. \end{aligned} \quad (34)$$

Here, the relation $(r/l)^{N/4} = (1 + av/D)^{N/4} \rightarrow \exp(\text{Pe}/4)$ has been used to evaluate the macroscopic limit. The asymptotically decaying density takes then the form

$$\rho(x, t) = \rho_B + \frac{\mathcal{A}}{L} (\mathcal{N}^{(t)} - \mathcal{N}^{(\infty)}) \exp\left(\text{Pe} \frac{2x - L}{4L}\right) \sin \frac{\pi x}{L}. \quad (35)$$

V. BOUNDARY CONTRIBUTIONS TO THE IRREVERSIBLE ENTROPY PRODUCTION

A. Absorbing boundaries

In the case of a long-term relaxation towards an empty state ($\rho_0 = \rho_{N+1} = \rho_B = 0$), Eq. (28) holds for the full density ρ_m , and one can write [see Eq. (22)]

$$\Sigma_t^{(\text{irr})} \equiv \kappa \mathcal{N} \exp(-\kappa\tau) = \kappa \mathcal{N}' \rightarrow \kappa \mathcal{N}, \quad (36)$$

where \mathcal{N}' is the number of particles at time $(n+1)\tau$.

The mixing term (24) can be expressed by means of the total entropy (19) to obtain

$$\Sigma_{\text{mix}}^{(\text{irr})} = \frac{1}{\tau}(e^{-\kappa\tau} - 1)S_{\text{tot}} \rightarrow -\kappa S_{\text{tot}}, \quad (37)$$

and in view of $\rho_0 = \rho_{N+1} = \rho_B = 0$ the boundary contribution (25) becomes

$$\begin{aligned} \Sigma_b^{(\text{irr})} &= -\frac{a}{\tau} \frac{\mathcal{AN}}{L} \sin \frac{\pi}{N+1} \left\{ r \left(\frac{r}{l} \right)^{(N-1)/4} \ln \left[\frac{\mathcal{AN}}{L\rho^*} \left(\frac{r}{l} \right)^{(N+3)/4} \sin \frac{\pi}{N+1} \right] \right. \\ &\quad \left. + l \left(\frac{r}{l} \right)^{-(N-1)/4} \ln \left[\frac{\mathcal{AN}}{L\rho^*} \left(\frac{r}{l} \right)^{-(N+3)/4} \sin \frac{\pi}{N+1} \right] \right\} \\ &= -\frac{a}{\tau} \frac{\mathcal{AN}}{L} \sin \frac{\pi}{N+1} \left\{ \left[r \left(\frac{r}{l} \right)^{(N-1)/4} + l \left(\frac{r}{l} \right)^{-(N-1)/4} \right] \ln \left[\frac{\mathcal{AN}}{L\rho^*} \sin \frac{\pi}{N+1} \right] \right. \\ &\quad \left. + \left[r \left(\frac{r}{l} \right)^{(N-1)/4} - l \left(\frac{r}{l} \right)^{-(N-1)/4} \right] \frac{N+3}{4} \log \frac{r}{l} \right\}. \quad (38) \end{aligned}$$

Observing that in the macroscopic limit both r and l are in leading order equal to $\tau D/a^2$, and that $\log(r/l) \rightarrow av/D$, one obtains for the relaxation contribution to the total irreversible entropy production

$$\Sigma_{\text{relax}}^{(\text{irr})} = \kappa \mathcal{N} \left[1 - \frac{\text{Pe}}{4} \tanh \frac{\text{Pe}}{4} - \ln \left(\frac{\mathcal{N}}{L\rho^*} \frac{\pi^2}{2} \frac{1 + (\text{Pe}/2\pi)^2}{\cosh(\text{Pe}/4)} \frac{a}{L} \right) \right] - \kappa S_{\text{tot}}. \quad (39)$$

This expression shows the expected logarithmic divergence since $(a/L) \rightarrow 0$ in the macroscopic limit. On the other hand, the result can properly be interpreted only after evaluating S_{tot} . In particular, the reference density ρ^* has to drop out again in the final result.

In the macroscopic limit the sum over m in the definition of S_{tot} becomes an integral. By using Eqs. (32) one finds

$$\begin{aligned} S_{\text{tot}} &= -\mathcal{AN} \int_0^1 dx \exp(-\text{Pe} \frac{2x-1}{4}) \sin(\pi x) \\ &\quad \ln \left[\frac{\mathcal{AN}}{L\rho^*} \exp \left(-\text{Pe} \frac{2x-1}{4} \right) \sin(\pi x) \right]. \quad (40) \end{aligned}$$

Applying the relation (34), we see that the specific entropy

$$S_{\text{tot}} = \mathcal{N} f \left(\text{Pe}, \frac{\mathcal{N}}{L\rho^*} \right) \quad (41)$$

is a function of the Péclet number and of the ratio of the average density in the system \mathcal{N}/L and the reference density ρ^* , *i.e.*, of another dimensionless constant that involves the parameter ρ^* selecting the origin of the

entropy scale (which, as mentioned earlier, is an arbitrary number in classical physics). The total entropy cannot be evaluated exactly. However, to obtain its behavior in leading order for very large and small Péclet numbers it is sufficient to approximate the expression under the logarithm by its maximum value. In the two limiting cases one thus finds in leading order in \mathcal{N}

$$S_{\text{tot}} = \begin{cases} -\mathcal{N} \ln \left(\text{Pe} \frac{\mathcal{N}}{\rho^* L} \right) & \text{for } \text{Pe} \gg 1, \\ -\mathcal{N} \ln \left(\frac{\mathcal{N}}{\rho^* L} \right) & \text{for } \text{Pe} \ll 1. \end{cases} \quad (42)$$

This implies for the specific entropy production

$$P^{(\text{irr})} = \frac{\Sigma_{\text{relax}}^{(\text{irr})}}{\mathcal{N}} = \begin{cases} \kappa \ln \left(\text{Pe}^{-1} \frac{L}{a} \right) & \text{for } \text{Pe} \gg 1, \\ \kappa \ln \frac{L}{a} & \text{for } \text{Pe} \ll 1. \end{cases} \quad (43)$$

The result shows the expected logarithmic divergence due to the boundary terms. It should be considered as an example clearly showing the inadequateness of global quantities for characterizing thermodynamic properties [*cf.* Eq. (1)]. The reason for the breakdown of the prediction (5) lies in the fact that the argument leading to this result focuses on the shrinking of the support of the measure by assuming the smoothness of the distribution along the unstable manifold. It thus entirely disregards that the density is very inhomogeneously distributed as a consequence of the absorbing boundaries. It should also be noted that the result obtained for the diffusive case $\text{Pe} \ll 1$ is the analog of the thermodynamic expression (1) since in this case $\rho = N\pi/(2L)\sin(x\pi/L)$, such that the parameter α of Eq. (1) takes the value $\alpha = \mathcal{N}\pi/(2L^2)$ and $D\alpha = \mathcal{N}\kappa/2$. The presence of factor 1/2 is due to the fact that Eq. (1) gives the contribution of one end only.

B. Influence of a small background density

We now assume that ρ_B is nonzero but much smaller than ρ_m except for a narrow boundary layer where the sine of (33) approaches zero. In that case Eq. (28) gives an upper bound to the ratio of densities at successive times, that is very accurate in the interior of the system. Consequently, the evaluation of (26) carries over except that the boundary term $\Sigma_b^{(\text{irr})}$ picks up contributions due to the finite density ρ_B in the cells 0 and $N+1$. In the macroscopic limit this term becomes

$$\Sigma_b^{(\text{irr})} \rightarrow -\kappa(\mathcal{N} - \mathcal{N}^{(\infty)}) \left(1 + \ln \frac{\rho_B}{\rho^*} \right) \approx -\kappa\mathcal{N} \left(1 + \ln \frac{\rho_B}{\rho^*} \right). \quad (44)$$

Since the total entropy S_{tot} and the contribution $\Sigma_t^{(\text{irr})}$ to the entropy production do not significantly change for a sufficiently small background density, the full entropy production becomes in the macroscopic limit

$$\Sigma_{\text{relax}}^{(\text{irr})} = -\kappa \mathcal{N} \ln \frac{\rho_B}{\rho^*} - \kappa S_{\text{tot}}. \quad (45)$$

After substituting S_{tot} from (42), we obtain the specific irreversible entropy production for $\mathcal{N} \gg \mathcal{N}^{(\infty)}$,

$$P^{(\text{irr})} = \begin{cases} \kappa \ln \left(\text{Pe}^{-1} \frac{\mathcal{N}}{\mathcal{N}^{(\infty)}} \right) & \text{for } \text{Pe} \gg 1, \\ \kappa \ln \left(\frac{\mathcal{N}}{\mathcal{N}^{(\infty)}} \right) & \text{for } \text{Pe} \ll 1. \end{cases} \quad (46)$$

The result clearly shows that the logarithmic divergences in the entropy production of the previous case are due to the vanishing of a physically indispensable background density ρ_B .

VI. DISCUSSION

The result (46) involves only well-behaved macroscopic quantities, and the logarithm of the ratio of the number \mathcal{N} of particles in the system over the number $\mathcal{N}^{(\infty)} = \rho_B L$ of particles approached in the steady state. At intermediate times, where \mathcal{N} is still much larger than $\mathcal{N}^{(\infty)}$, the ratio $\mathcal{N}/\mathcal{N}^{(\infty)}$ decreases to a good approximation exponentially like $\exp(-\kappa t)$ such that the rate of irreversible entropy production starts to decrease linearly like $-\kappa^2 t$. During this time regime the boundary contribution is by a factor of $\log(\mathcal{N}/\mathcal{N}^{(\infty)})$ larger than the bulk contributions accounted for by Eq. (5). Hence, even in the more realistic setting accounting for a finite background density, Eq. (5) only describes a sub-dominant contribution to the entropy-production rate. The reason for its failure is that the contributions arising from the spatial distribution of the particles and the induced inhomogeneous particle currents are not adequately taken into account by an entropy based solely on the conditionally-invariant measure.

In spite of the strong contributions due to the boundary terms, however, the entropy production remains proportional to the escape rate κ even in more general situations. Besides for the short times, where this condition follows from Eq. (46), this can be easily illustrated in the long-time regime, for which $(\mathcal{N} - \mathcal{N}^{(\infty)})\psi \equiv \Delta \mathcal{N} \psi \ll \rho_B$. Consequently, the entropy-production rate is obtained as

$$\begin{aligned} \Sigma^{(\text{irr})} &= \int dx \frac{\rho(x)}{D} \left(v - D \frac{\Delta \mathcal{N} \partial_x \psi}{\rho(x)} \right)^2 \\ &\approx \frac{v^2}{D} \int dx \rho(x) - \frac{\Delta \mathcal{N}^2}{\rho_B} D \int dx \psi(x) \partial_x^2 \psi(x) \end{aligned}$$

$$\approx \mathcal{N} \frac{v^2}{D} + \kappa \frac{(\Delta \mathcal{N})^2}{\mathcal{N}^{(\infty)}} g(\text{Pe}) \quad (47)$$

where $g(\text{Pe}) = \int dx \psi^2(x)$ is a function of Pe only. The corresponding irreversible entropy production due to relaxation is $\Sigma_{\text{relax}}^{(\text{irr})} = \Sigma^{(\text{irr})} - \mathcal{N} v^2/D$. In the second line in Eq. (47) the term proportional to $\partial_x \psi$ does not appear since its integral vanishes. Moreover, an integration by parts was used to obtain a second spatial derivative of the density which, according to the advection-diffusion equation, is proportional to its time-derivative, *i.e.*, it amounts to $-\kappa \psi$ for the slowest decaying mode ψ (again it is used here that terms proportional to $\partial_x \psi$ and $\psi \partial_x \psi$ vanish under the integral). The function $g(\text{Pe})$ can easily be evaluated for the multibaker map, but in general it depends on the shape of the system. Thus, the specific irreversible entropy production taken with respect to $\Delta \mathcal{N}$ is

$$P^{(\text{irr})} = \frac{\Sigma_{\text{relax}}^{(\text{irr})}}{\Delta \mathcal{N}} = \kappa \frac{\Delta \mathcal{N}}{\mathcal{N}^{(\infty)}} g(\text{Pe}). \quad (48)$$

Equation (48) implies that even in a general thermodynamic setting the relaxational entropy-production is proportional to the escape rate κ , which characterizes the approach towards the stationary state. In contrast to the dynamical-system arguments [3] based on the escape-rate formalism, the term involves in general a non-trivial function $g(\text{Pe})$ of the Péclet number, and it has an amplitude $\Delta \mathcal{N}/\mathcal{N}^{(\infty)}$ that is exponentially decaying like $\exp(-\kappa t)$.

It will certainly be interesting to investigate more closely the connection between the escape-rate formalism and the decay to systems supporting non-trivial stationary states. Another first step in this direction, which complements the present approach was suggested by Gilbert et al [32], who recently discussed the approach towards equilibrium in a system with periodic boundary conditions.

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